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**TITLE:** NEUTRON IRRADIATION OF  $\text{Nb}_3\text{Sn}$  AND  $\text{NbTi}$  MULTIFILAMENTARY COMPOSITES

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# NEUTRON IRRADIATION OF $\text{Nb}_3\text{Sn}$ AND $\text{NbTi}$ MULTIFILAMENTARY COMPOSITES\*

Don M. Parkin<sup>†</sup> and A. R. Sweedler<sup>‡</sup>

## ABSTRACT

$\text{NbTi}$  and  $\text{Nb}_3\text{Sn}$  multifilamentary composites have been irradiated with fast-neutrons at  $60 \pm 5^\circ\text{C}$  to fluences of  $1.2 \times 10^{19}$  n/cm<sup>2</sup> ( $E > 1$  MeV). The  $\text{NbTi}$  samples show only a moderate reduction of  $I_c$  as a function of neutron fluence in an applied field of 40 kG. Reductions in  $I_c$  were observed for fluences greater than  $3 \times 10^{17}$  n/cm<sup>2</sup> and saturate at 18% for fluences greater than  $3 \times 10^{19}$  n/cm<sup>2</sup>. The  $\text{Nb}_3\text{Sn}$  composites showed large neutron radiation induced changes in  $T_c$ ,  $I_c$  and  $H_{c2}$ . Reductions in  $T_c$  were observed for fluences greater than  $7 \times 10^{17}$  n/cm<sup>2</sup>. No measurable changes in  $I_c$  (40 kG) were observed below  $10^{18}$  n/cm<sup>2</sup>. Between 2 and  $3 \times 10^{18}$  n/cm<sup>2</sup>, however, there is an apparent threshold where a very rapid reduction in  $I_c$  (40 kG) is initiated. At the threshold the decrease in  $T_c$  is 13%. Between the threshold and  $1.1 \times 10^{19}$  n/cm<sup>2</sup>,  $I_c$  (40 kG) has been reduced to 4% of the unirradiated value. These changes in superconducting properties in  $\text{NbTi}$  and  $\text{Nb}_3\text{Sn}$  are analyzed in terms of the radiation induced defects. The impact of the response to irradiation of both materials on their applications in fusion reactor magnets is discussed.

## I. INTRODUCTION

Superconducting magnets have been proposed as integral components of new High Energy Accelerators and Controlled Thermonuclear Reactors (CTR's). Superconducting magnets in both applications will be subject to radiation fields in which the superconductor will undergo radiation damage. The most important of these applications in terms of sustained radiation flux, economics and impact on society is superconducting magnets for plasma confinement in fusion reactors. The large size, 10-20 meter bore for toroidal field coils, and the magnetic field at the plasma, 40 to 60 kG, are important parameters in determining the economics of a fusion reactor concept.<sup>1</sup> As a result, the response of technologically important superconductors to radiation exposure can play an important role in determining the magnet design characteristics and costs of a reactor concept.

The two superconductors most frequently considered for application are  $\text{NbTi}$  and  $\text{Nb}_3\text{Sn}$ . The maximum field at the conductor for  $\text{NbTi}$  applications would be 80-90 kG and 150-160 kG for  $\text{Nb}_3\text{Sn}$ . Studies of the effects of radiation using neutrons<sup>2-5</sup> and energetic charged particles<sup>6-10</sup> have shown that changes in  $T_c$ ,  $T$ , and  $H_{c2}$  occur in both materials. Of the two materials,  $\text{Nb}_3\text{Sn}$  which has the A-15 structure, is more sensitive to radiation than  $\text{NbTi}$ . One specific observation of these studies is that not only are the unirradiated superconducting properties such as critical current a function of sample preparation, but the observed radiation damage effects are also dependent on the initial state of the sample. To study the effects of neutron irradiation in superconducting magnets, engineering materials in the form of multifilamentary composites were used in this study.

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## II. EXPERIMENTAL

Samples of  $\text{NbTi}$  and  $\text{Nb}_3\text{Sn}$  multifilamentary composites and single phase chemical vapor deposited  $\text{Nb}_3\text{Sn}$  were irradiated in the fast-neutron flux of the Brookhaven High Flux Beam Reactor (HFBR). The temperature of the samples during irradiation was  $60 \pm 5^\circ\text{C}$ . Irradiations were performed up to fluences of  $1.2 \times 10^{20}$  n/cm<sup>2</sup> ( $E > 1$  MeV). The  $\text{NbTi}$  samples were commercial "Supercon 402" wires with 402 filaments of  $\text{NbTi}$  and a composite diameter of 0.02 cm. The  $\text{Nb}_3\text{Sn}$  samples were fabricated at Brookhaven National Lab. by M. Suenaga as 0.04 cm wire containing 19 filaments in a Cu-10 wt % Sn matrix. As shown in Fig. 1 the  $\text{Nb}_3\text{Sn}$  filaments consist of a copper core, unreacted Nb layer and a  $\text{Nb}_3\text{Sn}$  diffusion layer. A more detailed description of the sample material is given elsewhere.<sup>11</sup>

Critical current measurements as a function of applied field and measurement temperature were performed using a four-point resistance technique. The critical current was defined as the current for which 3 $\mu\text{V}$  was developed across a 1-cm length of sample. Each data point in the figures is the average value obtained from measurements on several independent samples. The critical temperature,  $T_c$ , measurements were made using a standard mutual inductance technique.

## III. DISCUSSION

**$\text{NbTi}$**  Results of the 4.2 K measurements of  $I_c$  on  $\text{NbTi}$  as a function of neutron fluence with  $H_J = 40$  kG are given in Fig. 2. Reductions in  $I_c$  (40 kG) were observed for fluences greater than  $3 \times 10^{17}$  n/cm<sup>2</sup>. At fluences above  $3 \times 10^{19}$  n/cm<sup>2</sup>, the reduction in  $I_c$  (40 kG) tends to saturate at 18%. For  $H_J$  less than 40 kG, however,  $I_c$

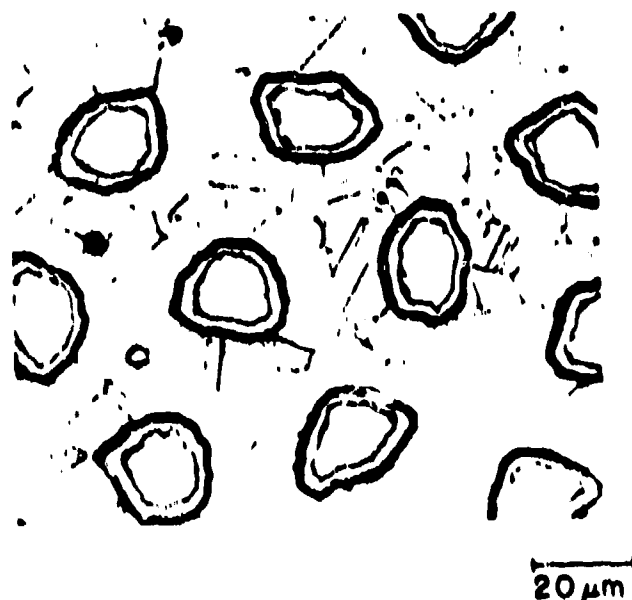


Fig. 1. Photomicrograph of a 19-filament  $\text{Nb}_3\text{Sn}$  composite wire. The central portion of each filament is copper. The unreacted niobium can be seen between the copper core and the darker  $\text{Nb}_3\text{Sn}$  border. The matrix is Cu-10 wt % Sn.

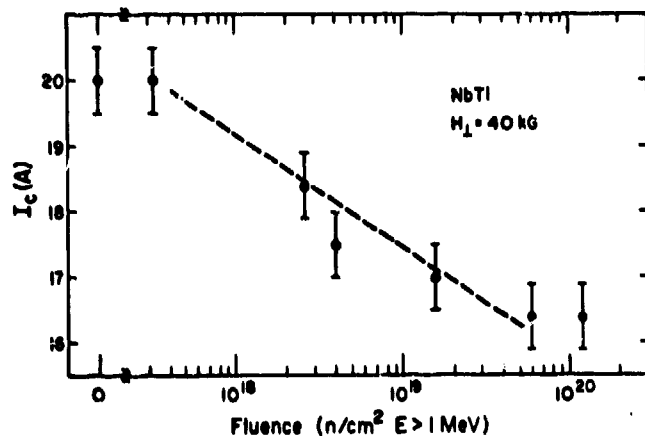


Fig. 2.  $I_c$  (40 kG) at 4.2 K as a function of fast-neutron fluence for NbTi multifilamentary composites.

continues to decrease. From  $1.6 \times 10^{19}$  to  $1.2 \times 10^{20}$  n/cm<sup>2</sup> the decrease in  $I_c$  at 10 and 20 kG goes from 21 and 17% to 29 and 27% respectively. Further, for  $H_1$  less than 40 kG, the  $I_c$  reductions are not only larger but are observed at lower fluences than at 40 kG.

The magnitude of the reduction in  $I_c$  (40 kG) can be compared to other neutron irradiation studies of NbTi filaments. Soell et al.<sup>12</sup> irradiated bare filaments at 5 K with neutrons. After irradiation they annealed their samples at room temperature. After annealing a 15% reduction in  $I_c$  remained at a fluence of  $7.5 \times 10^{18}$  n/cm<sup>2</sup>. The estimated reduction in  $I_c$  from Fig. 2 for this study is 12%.

Decreases in  $I_c$  in NbTi have been observed after low temperature neutron irradiation and decreases in  $I_c$ ,  $T_c$  and  $H_{c2}$  have been observed after low temperature proton<sup>10</sup> and deuteron<sup>11</sup> irradiation. The general model which these authors have used to analyze the observed reductions in  $I_c$  is one in which the radiation induced defects reduce the pinning strength of preexisting pinning centers. The pinning strength is reduced by the decreasing difference in defect concentration between that in the matrix and that in the pinning centers. Wohleben<sup>1</sup> has extended this analysis to include the effects of reductions in  $T_c$  and  $H_{c2}$ . He concludes that after low temperature proton irradiation 70% of the decrease in  $I_c$  arises from a reduction in pinning strength and 30% from a reduction in  $T_c$  or  $H_{c2}$ . In the present study only decreases in  $I_c$  were measured so that a detailed application of Wohleben's model can not be made. Wohleben observed a constant percentage degradation of the  $I_c(H_1, T_m)$  surface for the  $3 \times 10^{16}$  and  $1 \times 10^{17}$  p/cm<sup>2</sup> irradiations and a 7.7% reduction in  $I_c$  was found after the  $1 \times 10^{17}$  p/cm<sup>2</sup> irradiation plus room temperature anneal. This data can be compared to the present data by calculating the radiation induced defect production cross section for neutrons and protons. Approximate calculations of the defect production cross sections show that the proton data includes the defect production range  $10^{-1}$  to  $3 \times 10^{-3}$  dpa and the present neutron data  $10^{-6}$  to  $10^{-1}$  dpa. In the same dpa range the two sets of data show differing radiation effects. In the neutron data,  $I_c(H_1, T_m)$  is not constant and the corresponding reduction in  $I_c$  when compared to the annealed

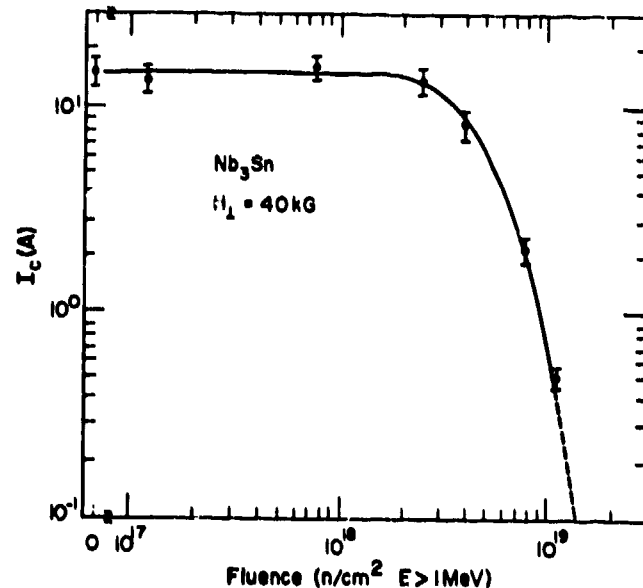


Fig. 3.  $I_c$  (40 kG) at 4.2 K as a function of fast-neutron fluence for Nb<sub>3</sub>Sn multifilamentary composites.

proton data are larger in the neutron case. These differences could be related to the type of irradiation and the sample material.

#### Nb<sub>3</sub>Sn

Large reductions in  $T_c$ ,  $H_{c2}$  and  $I_c$  were observed in Nb<sub>3</sub>Sn after neutron irradiation. This sensitivity to irradiation has also been observed by others after neutron<sup>2-5</sup> proton,<sup>6,8,10</sup> deuteron<sup>7</sup> and Oxygen<sup>9</sup> bombardment. Little or no reduction in  $I_c$  for  $H_1 \leq 40$  kG was observed for neutron fluence below  $10^{18}$  n/cm<sup>2</sup>. Between 2 and  $3 \times 10^{18}$  n/cm<sup>2</sup>, however, there is an apparent threshold where a very rapid reduction in  $I_c$  (40 kG) is initiated as seen in Fig. 3. Between the threshold and  $1.1 \times 10^{19}$  n/cm<sup>2</sup>  $I_c$  (40 kG) has been reduced to 4% of the unirradiated value.

The response of  $T_c$  to neutron irradiation is shown in Fig. 4. The normalized critical temperature,  $T_n = T_c(\phi t)/T_c(0)$  is compared for the Nb<sub>3</sub>Sn composite and single phase Nb<sub>3</sub>Sn. The initial  $T_c$ 's are 15 and 18.1 respectively. Measurements of  $T_c$  on the Nb<sub>3</sub>Sn

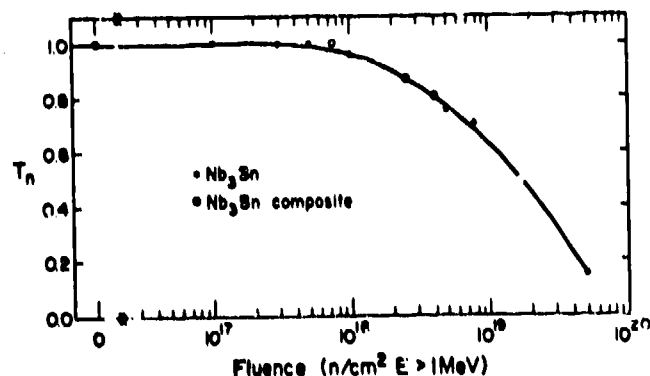


Fig. 4. Normalized critical temperature  $T_n = T_c(\phi t)/T_c(0)$  as a function of fast-neutron fluence for Nb<sub>3</sub>Sn and Nb<sub>3</sub>Sn multifilamentary composites.

\* dpa is a radiation damage unit which means displacements/atom and is a measure of the number of displacements or interstitial-vacancy pairs produced per lattice atom after a given time integrated flux or fluence.<sup>11</sup>

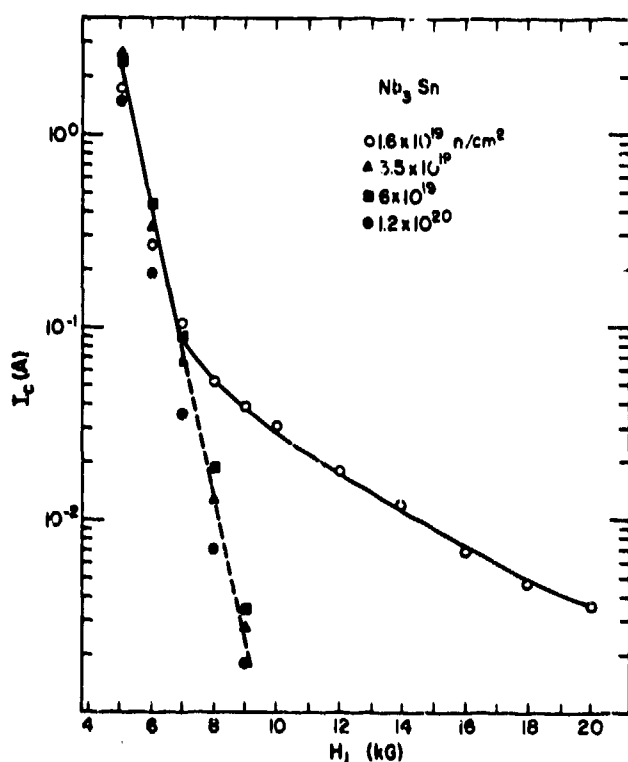


Fig. 5.  $I_c$  at 4.2 K as a function of  $H_1$  for  $Nb_3Sn$  multifilamentary composites irradiated with fast-neutrons from  $1.6 \times 10^{19}$  to  $1.2 \times 10^{20}$   $n/cm^2$ .

composite could only be made for  $T_c$  greater than 9.1 K, the  $T_c$  for the Nb cores in the composite. In this temperature range the normalized reduction in  $T_c$  agrees with that for the pure  $Nb_3Sn$ . At the current threshold the  $T_c$  reduction is 13%.

The magnitude of the radiation induced reductions in  $H_{c2}$  (4.2 K) are indicated in Figs. 1 and 5. Between  $1.1 \times 10^{19}$  and  $1.6 \times 10^{19}$   $n/cm^2$   $H_{c2}$  (4.2 K) is reduced below 40 kG. In this fluence range the projected  $T_c$  changes from ~ 8.9 to ~ 7.5 K.

Measurements of  $I_c$  as a function of  $H_1$  to 40 kG and measuring temperature,  $T_m$  to 1.9 K, were conducted on samples irradiated to fluences greater than  $1.6 \times 10^{19}$   $n/cm^2$ . The results of these measurements are summarized by the data in Fig. 5. Within the data scatter indicated in Fig. 5 no fluence dependence of  $I_c(H_1, T_m)$  could be detected in these samples. Further the scatter in the data was not a systematic function of fluence. The influence of  $H_{c2}$  and  $T_c$  on  $I_c$  was expected to be measurable since the projected  $T_c$ 's are from 0-4 K. The results of these measurements can be interpreted in two ways: 1. in the  $Nb_3Sn$  composite, the fluence dependence of  $T_c$  is greater than for pure  $Nb_3Sn$  and  $T_c$  was below 1.9 K for all samples, 2. for all  $T_m$  and  $H_1$ ,  $H_{c2}(Nb_3Sn)$  was less than for the Nb cores and  $I_c(H_1, T_m)$  for  $Nb_3Sn$  was much less than for the Nb cores making the  $Nb_3Sn$  properties not detectable in the presence of the Nb. It was not possible to determine which interpretation or combination of the two is correct. In either case, the measurements are believed to show the properties of the Nb cores. This interpretation is supported by the fact that an  $H_{c2}$  (4.2 K) = 9 kG is in agreement with the value of 9.3 kG reported by Keil et al.<sup>15</sup> for annealed Nb.

Increases followed by decreases in  $I_c$  in  $Nb_3Sn$  as a function of irradiation fluence have been reported in

the literature.<sup>8,9</sup> The basic model used to analyze this data is one in which the radiation induced defects act as pinning centers which enhance the current and after a critical concentration is reached their pinning effectiveness is reduced by further irradiation and the current decreases. The amount of  $I_c$  increase being a function of initial  $I_c$  value and type of irradiation. In the present results no  $I_c$  increase was observed, suggesting that the radiation induced pinning centers were not as effective as the preexisting ones in the composite samples. However, the present data have two important similarities with the literature data. The fluence dependence of the reduction in  $I_c$  in the composite samples is similar to the fluence dependence of the decrease in  $I_c$  after the peak increase observed by others. This is especially true of the oxygen irradiation data. Second, the point at which  $I_c$  starts to decrease in both cases occurs at the same value of dpa, i.e.  $\approx 10^{-3}$  dpa. Stated in another way, a rapid decrease in  $I_c$  occurs after about the same number of defects have been produced in  $Nb_3Sn$  independent of sample preparation or type of irradiation.

At the point where the decrease in  $I_c$  occurs, there is a corresponding 13% reduction in  $T_c$ . The reduction in  $T_c$  following neutron irradiation in  $Nb_3Sn$  have been shown to be a general phenomenon in a large number of Nb based A-15 superconductors.<sup>16,17</sup> The observed changes in  $T_c$  have been correlated with radiation induced disorder in the A-15 structure. When an atom moves through an ordered lattice such as that in  $Nb_3Sn$  after receiving kinetic energy from a neutron scattering event, both point defects and lattice disorder are produced as its energy is dissipated.

In  $Nb_3Sn$  the superconducting properties are altered both by radiation induced point defects and radiation induced disorder. At the start of the rapid decrease in  $I_c$ , the lattice contains not only a sufficient concentration of radiation induced interstitial and vacancy type defects that can directly effect  $I_c$ , but also enough radiation induced disorder to produce large reductions in  $T_c$ . Both of these types of radiation damage must be considered in analyzing the effect of radiation on  $I_c$  in  $Nb_3Sn$ . The radiation induced disorder and the resultant changes in  $T_c$  and  $H_{c2}$  must be incorporated in a model that describes the changes in  $I_c$ . The radiation induced change in  $I_c$  in  $Nb_3Sn$  are due to flux pinning effects and significant changes in the fundamental superconductivity parameters.

#### IV. CONCLUSIONS

Neutron irradiation at  $60 \pm 5^\circ C$  of  $NbTi$  multifilamentary composites shows only moderate reductions in  $I_c$  (40 kG). Reductions in  $I_c$  were observed for fluences greater than  $3 \times 10^{17}$   $n/cm^2$  and saturation at 16% was observed for fluences above  $3-4 \times 10^{19}$   $n/cm^2$ . The present neutron irradiation data were compared to other irradiation data. It was observed that the present data agreed with low temperature neutron irradiation data but some differences with low temperature proton data were discussed. The observed reductions in  $I_c$  are primarily caused by a radiation induced reduction in pinning strength. Measurements of  $T_c$  and  $H_{c2}$  were not made so that an analysis of the effects of reductions in  $T_c$  and  $H_{c2}$  on  $I_c$  could not be made.

Large reductions in  $T_c$ ,  $H_{c2}$  and  $I_c$  were observed in the  $Nb_3Sn$  multifilamentary composites. A very rapid decrease in  $I_c$  (40 kG) occurs for neutron fluences greater than  $2-3 \times 10^{18}$   $n/cm^2$ . Measurable decreases in  $T_c$  were observed for fluences greater than  $7 \times 10^{17}$   $n/cm^2$ . Reductions in  $H_{c2}$  were also observed. The sensitivity of  $Nb_3Sn$  to irradiation is due to radiation induced defects and radiation produced disorder in the A-15 structure.

The operating parameters for superconducting magnets in application in radiation fields such as in a fusion reactor will be at temperatures near 4.2 K and at maximum fields at the conductor of  $\approx 160$  kG for  $\text{Nb}_3\text{Sn}$ , and 90 kG for  $\text{NbTi}$ . Since reductions in  $T_c$  and  $H_{c2}$  have been observed for both  $\text{NbTi}$  and  $\text{Nb}_3\text{Sn}$  it is reasonable to speculate that the radiation induced degradation of the superconducting properties might be greater and occur at lower fluences than under the conditions discussed in this paper. The relative magnitude of the radiation effects discussed in this paper suggest that  $\text{Nb}_3\text{Sn}$  would be more sensitive to irradiation under these conditions than  $\text{NbTi}$ .

#### REFERENCES

1. For a review of current fusion reactor parameters see papers in Proc. Fifth Symposium on Engineering Problems of Fusion Research, Princeton, 1973 (IEEE, New York, 1974) and Proc. First Topical Meeting on the Technology of Controlled Nuclear Fusion, San Diego, 1974 (USAEC, CONF-740402).
2. G. W. Cullen, R. L. Novak, and J. P. McEvoy, Jr., *JCA Rev.* **25**, 479 (1964).
3. J. P. McEvoy, R. F. Decell, and R. L. Novak, *Appl. Phys. Lett.* **4**, 43 (1964).
4. P. S. Swartz, H. R. Hart, Jr., and R. L. Fleisher, *Appl. Phys. Lett.* **4**, 71 (1964).
5. G. W. Cullen and R. L. Novak, *J. Appl. Phys.* **37**, 3348 (1966).
6. H. F. Bode and K. Wohleben, *Phys. Lett.* **A24**, 25 (1967).
7. H. T. Coffey, E. L. Keller, A. Patterson, and S. H. Autler, *Phys. Rev.* **155**, 358 (1967).
8. K. Wohleben, *Z. Angew. Phys.* **27**, 52 (1969).
9. G. Ischenko, H. Mayer, H. Voit, B. Besslein, and E. Handl, *Z. Physik*, **256**, 176 (1972).
10. K. Wohleben, *J. Low Temp. Phys.* **13**, 269 (1973).
11. D. M. Parkin and D. G. Schweitzer, *Nuclear Technology* **22**, 108 (1974).
12. M. Soell, S. L. Wiff, and G. Vogl in Proc. Conf. Applied Superconductivity, Annapolis, 1972 (IEEE, New York, 1972), p. 434.
13. D. G. Doran, *Nucl. Sci. Eng.* **49**, 130 (1972).
14. R. Bett, AERE, Harwell, unpublished.
15. D. Keil, U. Merbold, and J. Diehl, *Appl. Phys.* **3**, 217 (1974).
16. A. R. Sweedler, D. G. Schweitzer, and G. W. Webb, to be published in *Phys. Rev. Lett.*
17. A. R. Sweedler, D. Cox, D. Schweitzer, and G. W. Webb, this conference.